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DETERMINATION OF THE THORON DAUGHTER WORKING LEVEL BY A ONE GROSS ALPHA-COUNT

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DETERMINATION OF THE THORON DAUGHTER WORKING LEVEL BY A ONE GROSS ALPHA-COUNT

by

J. Bigu* and W.K. Lau**

ABSTRACT

A study has been done on the determination of the thoron daughter Working Level, WL(Tn), by a one gross α-count. The relationship between the gross α-count rate per unit of volume of air sampled and WL(Tn), denoted the F-factor, has been investigated as a function of sampling time, elapsed time from the end of the sampling period, i.e., waiting time, and the thoron daughter disequilibrium ratio [ThC]/[ThB]. It has been found that F depends on both the waiting time and [ThC]/[ThB]. If α-count measurements are made at least 300 min after the end of sampling, F changes by less than 10% over the full range of theoretical values of [ThC]/[ThB], i.e., from 0 to 1. The F-factor is independent of [ThC]/[ThB] at approximately 215 min after the end of sampling. This feature can be used to determine WL(Tn) with higher accuracy and at least 1.5 hr earlier than is commonly done using other one gross α-count methods reported in the literature.

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INTRODUCTION

Extended inhalation of short-lived decay products of radon, i.e., radon daughters, poses a potential risk to human health. Because of this, monitoring of radon daughter levels in air for dose exposure calculation purposes is mandatory in uranium mines.

Until recently, radon daughters were the only airborne radioisotopes of concern from the health physics standpoint in uranium mines. However, experimental and theoretical studies have indicated that attention should also be paid to the short-lived decay products of thoron (thoron daughters) and to long-lived radioisotopes associated with dust in the respirable range.

Concern regarding the thoron daughter levels in uranium mines has been expressed because in certain mines (e.g., Alaska and Ontario) these levels are substantially higher than originally anticipated. Since no regular monitoring of thoron daughters in mines is normally carried out, there is in general some uncertainty regarding the extent of the airborne thoron daughter contribution in uranium mines.

Because thorium compounds are frequently found along with uranium minerals, careful mine air monitoring is likely to show that the presence of thoron daughters is a general problem rather than an isolated or particular one.

There is a variety of methods available for the determination of radon daughter levels (1 to 20). Comparatively speaking, however, little work has been done in developing methods for the determination of thoron daughter levels or radon daughters and thoron daughters mixtures (21 to 25).

The simplest method for determining the thoron daughter Working Level, WL(Tn), is by the one gross α-count method investigated by Rock (25). This method is essentially the same as that developed by Kusnetz for radon daughters (2,3). The method consists of sampling air for a given period of time through

an absolute filter by means of a pump of known flow-rate. The α-activity arising from the decay products of thoron is subsequently measured at a pre-established time after the end of the sampling period by means of an adequate detector/scaler arrangement.

The work presented here pertains to a study on the determination of the Working Level of thoron daughters by the one gross α-count method, and is an extension of previous work published elsewhere. The data are useful with regards to the thoron calibration facility at the Elliot Lake Laboratory, (CANMET, Energy, Mines and Resources Canada).

THEORETICAL BACKGROUND

WL(Tn) can be determined from a one gross α-count according to (25):

$$WL(Tn) = A/F \qquad \text{Eq 1}$$

where A represents the absolute α-disintegration rate from the α-emitters deposited in the filter during the sampling period per unit of air volume sampled. F is a conversion factor that can be determined experimentally and theoretically. A in Equation 1 can be expressed as:

$$A = N_\alpha/\varepsilon\eta\delta VT_c \qquad \text{Eq 2}$$

N_α in the above expression represents the number of α-counts recorded by an α-detector/scaler assembly with α-counting efficiency ε during the counting time T_c. The symbol $\eta(<1)$ is introduced to take into account absorption of α-particles in the filter, i.e., self-absorption, and other filter effects. The symbol $\delta(<1)$ is introduced to take into account plate-out of decay products in the sampling head. V is the volume of air sampled. If T_s and Q are, respectively, the sampling time (min) and sampling flow-rate (L/min), one can write: $V = QT_s$.

Equation 1 shows that WL(Tn) can easily be determined if F is known

as the other variables are determined experimentally. The conversion factor F can be determined from independent measurements of WL(Tn) by a different method to the one investigated here, or alternatively, it can be calculated analytically, as indicated below.

$$F = A_{theor}/WL(Tn)_{theor} \qquad \text{Eq 3}$$

The subscript theor in the above expression stands for theoretical. A_{theor} is given by an expression similar to Equation 2 where N_α is calculated theoretically, i.e., $N_{\alpha,theor}$. In order to calculate $N_{\alpha,theor}$, the following simplifying assumptions have been made:

1. The thoron daughter concentration in air remains constant during the sampling period.
2. The sampling pump flow-rate is constant during the sampling period.
3. The efficiency of counting is not energy dependent. This is not strictly true, but an average value over the energy range of interest can be taken by calculating the α-detector with standard radon daughter and thoron daughter sources for the filter/detector geometry of interest.

The rate of growth of activity on the filter during sampling is given by the following expression:

$$\frac{dA_m}{dt} = C_m Q + \lambda_m A_{m-1} - \lambda_m A_m \qquad \text{Eq 4}$$

where, A_m and A_{m-1} are the activities of the m-th and (m-1)-th members of the radioactive decay chain on the filter (pCi or Bq), respectively;

C_m is the atmospheric concentration of the m-th member of the radioactive decay chain (pCi/L or Bq/L);

λ_m is the decay constant of the m-th member of the radioactive decay chain (min^{-1});

The daughters activities accumulated on the filter between times T_{w1} and T_{w2} after the end of the sampling period for a sampling period T_s, IAW,

are given by (22,26):

$$IA_iW = QC_i\ \tau_i^2\ f_i \qquad \text{Eq 5}$$

$$IA_jW = Q\left[C_i\left(\frac{\tau_i^3 f_i}{\tau_i-\tau_j} + \frac{\tau_j^2\tau_i f_j}{\tau_j-\tau_i}\right) + C_j\ \tau_j^2\ f_j\right] \qquad \text{Eq 6}$$

$$IA_kW = Q\left[C_i\left(\frac{\tau_i^4 f_i}{(\tau_i-\tau_j)(\tau_i-\tau_k)} + \frac{\tau_j^3\tau_i f_j}{(\tau_j-\tau_i)(\tau_j-\tau_k)} + \frac{\tau_k^3\tau_i\ f_k}{(\tau_k-\tau_i)(\tau_k-\tau_j)}\right) + C_j\left(\frac{\tau_j^3 f_j}{\tau_j-\tau_k} + \frac{\tau_k^2\tau_j f_k}{\tau_k-\tau_j}\right) + C_k\ \tau_k^2\ f_k\right] \qquad \text{Eq 7}$$

where,

$$f_i = (1 - e^{-T_S})\ (e^{-T_{w1}/\tau_i} - e^{-T_{w2}/\tau_i}) \qquad \text{Eq 8}$$

$$f_j = (1 - e^{-T_S})\ (e^{-T_{w1}/\tau_j} - e^{-T_{w2}/\tau_j}) \qquad \text{Eq 9}$$

$$f_k = (1 - e^{-T_S})\ (e^{-T_{w1}/\tau_k} - e^{-T_{w2}/\tau_k}) \qquad \text{Eq 10}$$

As the above expressions are valid for both radon daughters and thoron daughters, the subindices i, j and k stand, respectively, for RaA or ThA, RaB or ThB and RaC or ThC. The symbol $\tau(= 1/\lambda)$ is the mean life of the radioisotope of interest.

For thoron daughters, IA_jW needs not to be considered as ThB is a β-emitter. Furthermore, the half-life of ThA is so short (i.e., 0.16 sec) that no appreciable α-particle contribution from this radioisotope can be measured. Hence, only the contribution from ThC, i.e., IA_kW, needs to be considered. However, the first term in round brackets of Equation 7 is very small compared with terms containing C_j and C_k. Thus, Equation 7 may be simplified to:

$$IA_kW \simeq QC_j\left[\left(\frac{\tau_j^3 f_j}{\tau_j-\tau_k} + \frac{\tau_k^2\tau_j f_k}{\tau_k-\tau_j}\right) + R\tau_k^2\ f_k\right] \qquad \text{Eq 11}$$

$$= QC_jP(R) \qquad \text{Eq 12}$$

where P(R), a function of R, is the term in square brackets in Equation 11 and $R = C_k/C_j$ is the thoron daughter disequilibrium ratio.

The theoretical number of α-counts, $N_{\alpha,theor}$, registered by an

α-detector/scaler assembly of counting efficiency ε corresponding to a sampling time T_s during the counting period from T_{w1} to T_{w2} can be written in terms of Equations 11 and 12 as follows:

$$N_{\alpha,theor} = (2.22)\,\varepsilon\eta\delta\, IA_kW \qquad \text{Eq 13}$$

The numerical value in round brackets, i.e., 2.22, is a conversion factor to convert activity in pCi into disintegrations per minute (dpm). If the filter activity is given in dpm, the above numerical factor should be deleted. (Note: for simplicity in what follows the symbols C_i, C_j and C_k, corresponding to ThA, ThB and ThC, will be substituted by [ThA], [ThB] and [ThC], respectively).

The general expression for the Working Level, WL, for radon daughters or thoron daughters is given as:

$$WL = \Sigma_m WL_m = 1.71 \times 10^{-5} \Sigma_m\, C_m\, \tau_m\, E_m\,(\alpha) \qquad \text{Eq 14}$$

$E_m(\alpha)$ is the ultimate α-particle energy (MeV) per decaying atom of m-type. The numerical factor 1.3×10^5 (MeV) represents the total ultimate α-particle energy corresponding to the decay of an initial mixture of 100 pCi/L (3.7 Bq/L) each of RaA, RaB, RaC and RaC' (for radon daughters) or 7.5 pCi/L (0.277 Bq/L) each of ThB and ThC (for thoron daughters) in radioactive equilibrium with 100 pCi/L (3.7 Bq/L) of radon or 7.5 pCi/L (0.277 Bq/L) of thoron, respectively. (Note the condition of radioactive equilibrium is not necessary, but the ultimate α-energy 1.3×10^5 MeV/L is.)

For thoron daughters, Equation 14 becomes:

$$WL(Tn) = 0.1222[ThB] + 0.0116[ThC]$$

$$= [ThB]\,(0.1222 + 0.0116R) \qquad \text{Eq 15}$$

From Equation 3, and Equations 2, 12, 13 and 15 one can easily show after some algebraic manipulation that:

$$F = \frac{P(R)}{T_s\, T_c\, (0.1222 + 0.0116R)} \qquad \text{Eq 16}$$

where $T_C = T_{w2} - T_{w1}$.

Equation 16 shows that the conversion factor is a function of the sampling period, the waiting times T_{w1} and T_{w2}, which also determine the counting period, and the thoron daughter disequilibrium ratio R = [ThC]/[ThB].

Equation 16 has been used to compute the conversion factor F as a function of T_s, T_{w1}, T_{w2} and R. The results of this calculation are shown in Fig. 1 to 6.

RESULTS AND DISCUSSION

Figures 1 and 2 show the theoretical F factor versus the ratio [ThC]/[ThB] for a sampling time of 5 min and 20 min, respectively, for several waiting times ranging from 400 min to 600 min. These figures show that F decreases with increasing waiting time and increasing ratio [ThC]/[ThB]. Furthermore, there is a linear relationship between F and [ThC]/[ThB]. However, the variation of F, i.e., ΔF, over the limiting theoretical values for [ThC]/[ThB], i.e., 0 and 1, is under 10%. The small value for ΔF is to be expected from examination of Equation 15 as the contribution from the ThB-containing term largely dominates the ThC term.

From Figures 1 and 2 it may also be inferred that for a given [ThC]/[ThB], F varies linearly with the waiting time. Comparison of data indicates that there is essentially no difference between a sampling time of 5 min and 20 min, two arbitrarily chosen, although convenient, times.

Figures 3 and 4 show similar data as those corresponding to Fig. 1 and 2, but for waiting times ranging from 0 min to 200 min. The figures indicate that F is highly dependent on [ThC]/[ThB] and the waiting time. The difference in F becomes larger at lower values of [ThC]/[ThB] and shorter waiting times.

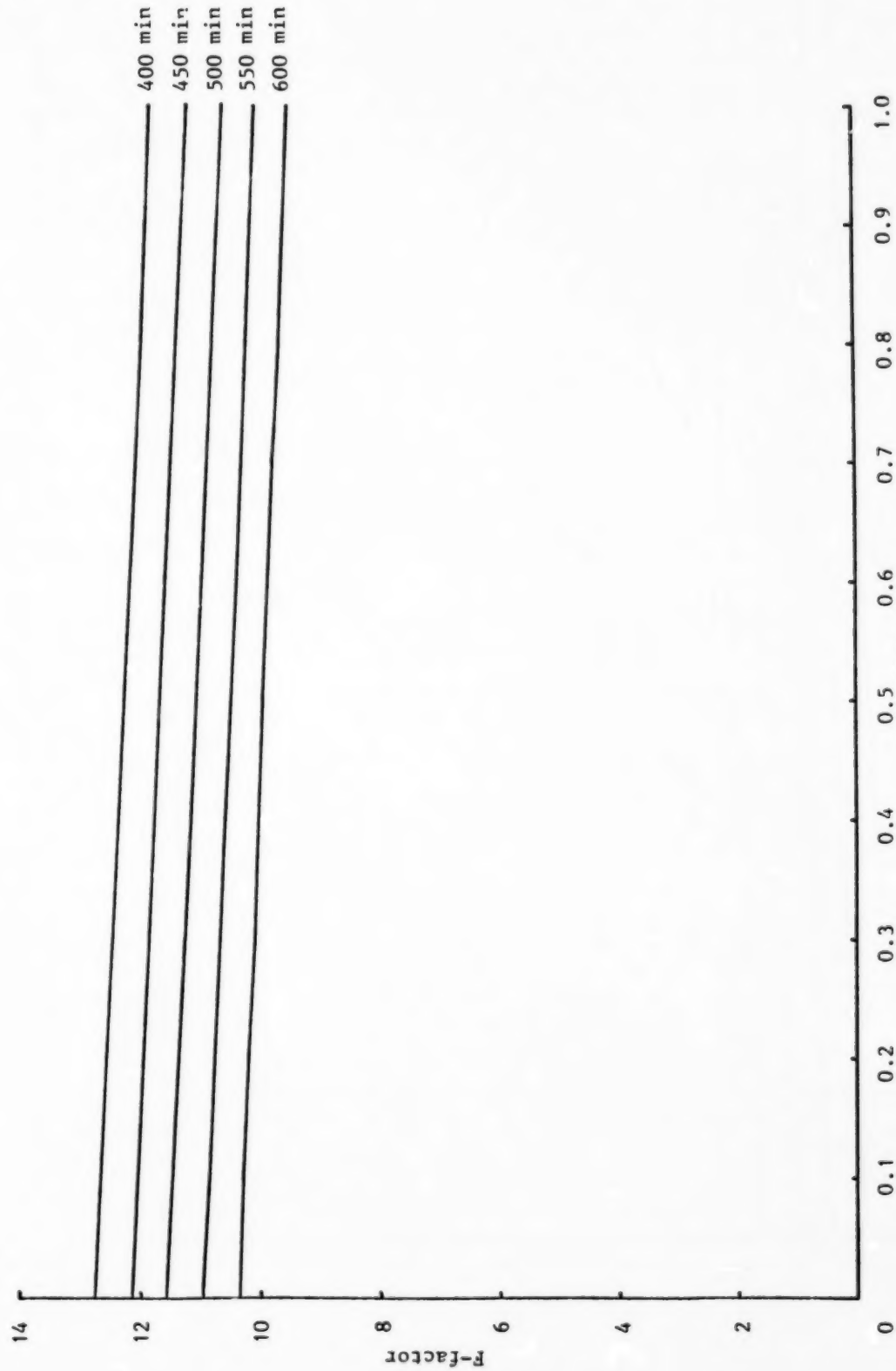

Fig. 1 - Theoretical F-factor versus the ratio [ThC]/[ThB] for the following waiting times (T in min): 400, 450, 500, 550 and 600. Sampling time is 5 min.

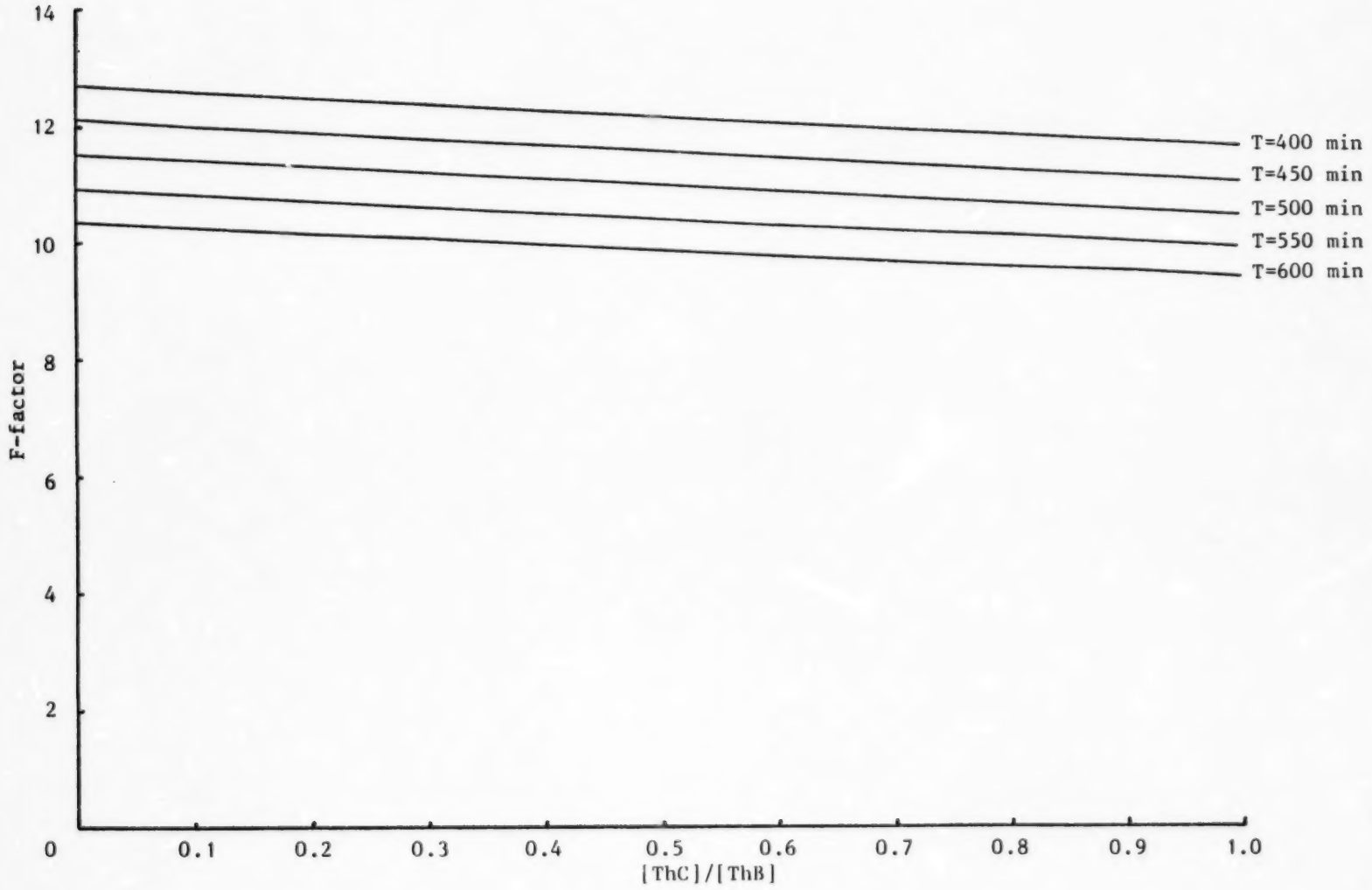

Fig. 2 - Theoretical F-factor versus the ratio [ThC]/[ThB] for the following waiting times (T in min): 400, 450, 500, 550 and 600. Sampling time is 20 min.

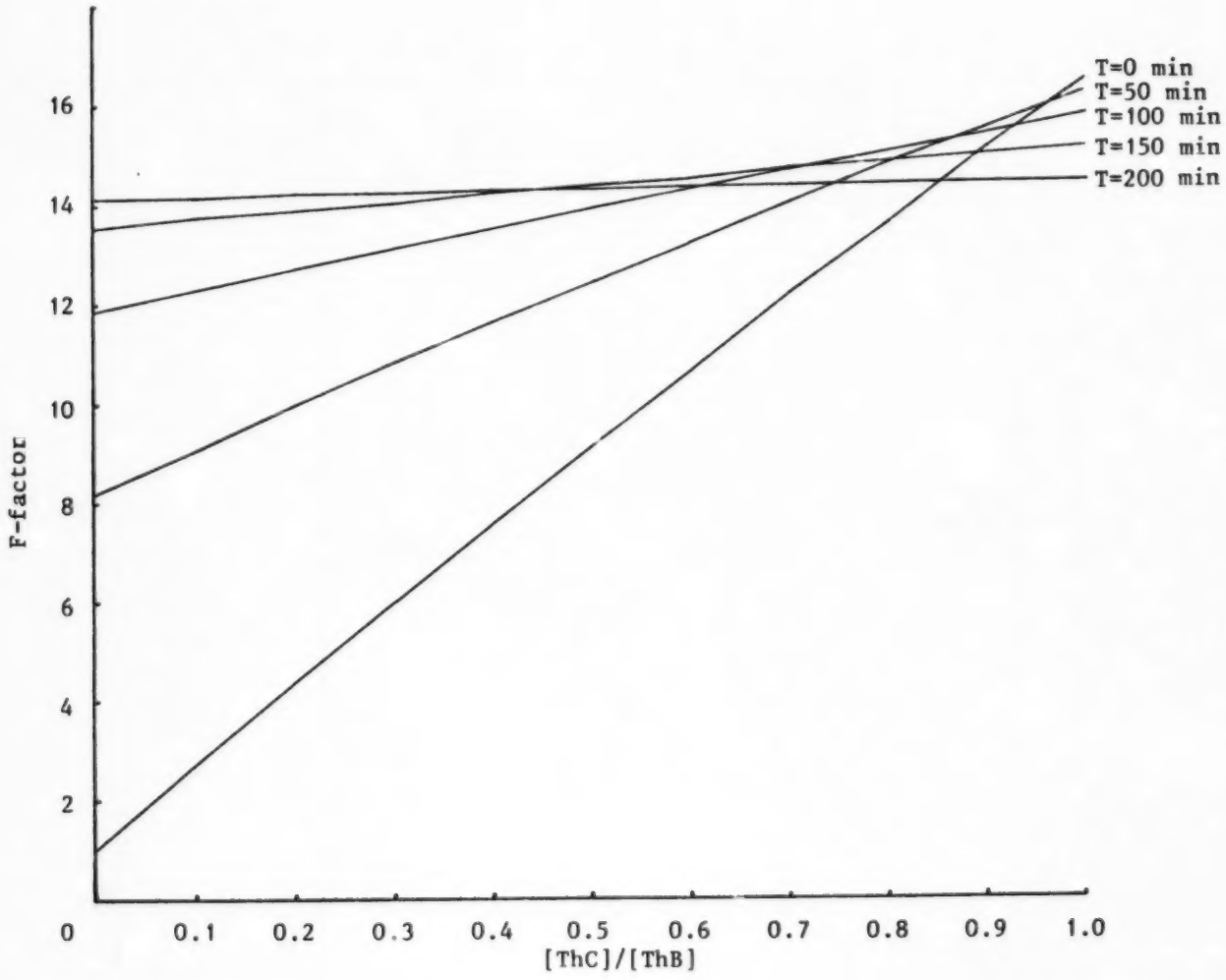

Fig. 3 - Theoretical F-factor versus the ratio [ThC]/[ThB] for the following waiting times (T in minutes): 0, 50, 100, 150 and 200. Sampling time is 5 minutes.

Fig. 4 - Theoretical F-factor versus the ratio [ThC]/[ThB] for the following waiting times (T in minutes): 0, 50, 100, 150 and 200. Sampling time is 20 minutes.

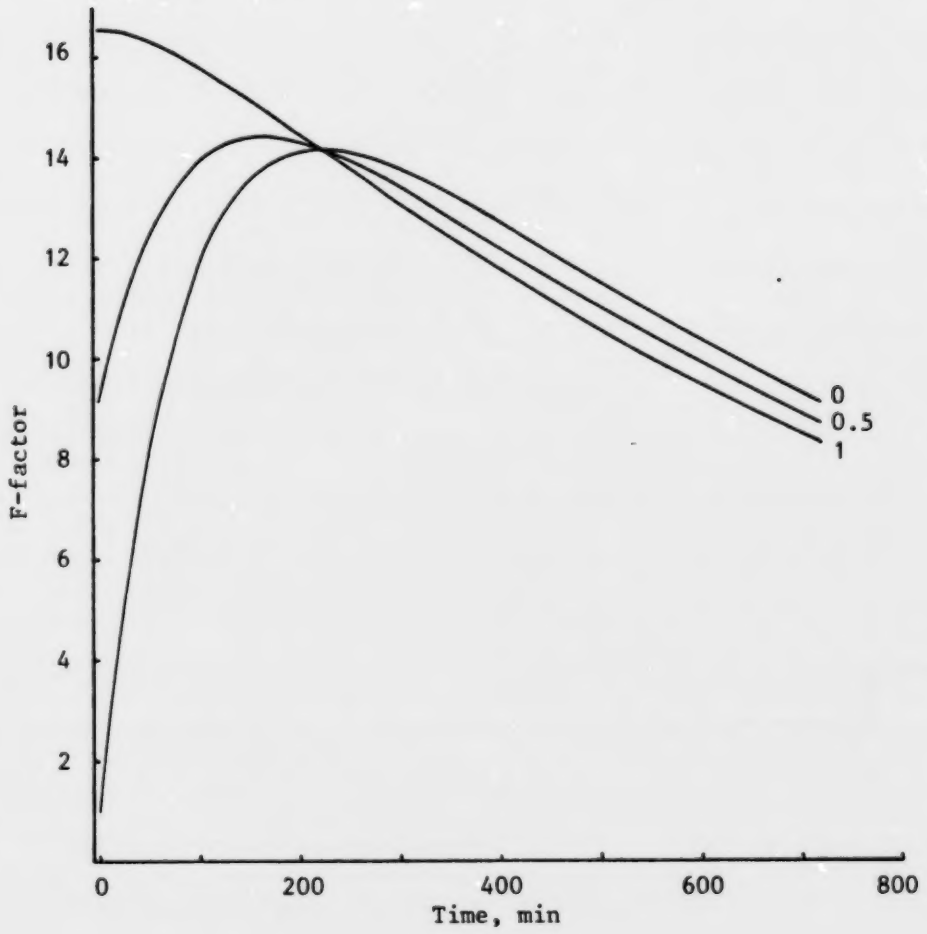

Fig. 5. - F-factor versus waiting time for the following [ThC]/[ThB] ratios: 0, 0.5 and 1. Sampling time is 5 minutes.

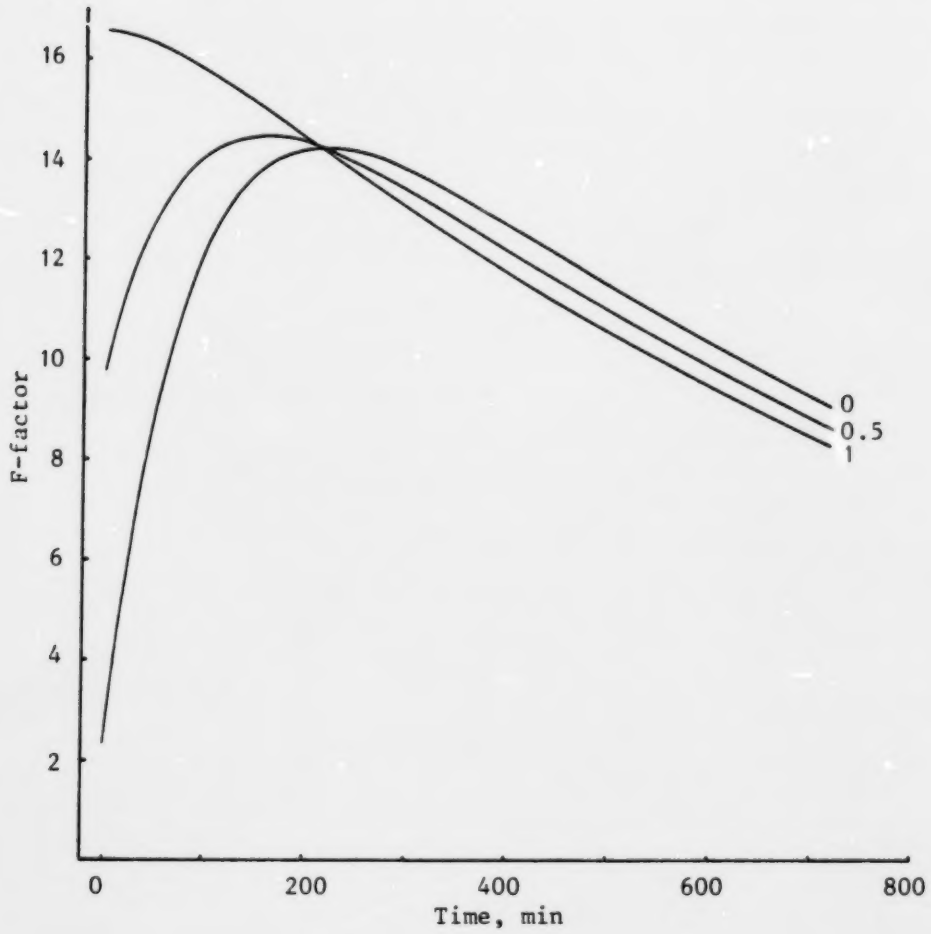

Fig. 6. - F-factor versus waiting time for the following [ThC]/[ThB] ratios: 0, 0.5 and 1. Sampling time is 20 min.

A comparison of Fig. 3 and 4 show that the value of F is lower for the shorter sampling time, i.e., 5 min, and that the difference becomes smaller with increasing values of [ThC]/[ThB]. Much less variation in F is observed at longer waiting times, a fact which is consistent with the data shown in Fig. 1 and 2.

Figures 5 and 6 show the F-factor versus the waiting time for a sampling time of 5 min and 20 min, respectively, for several values of the ratio [ThC]/[ThB]. These figures indicate that there is a strong dependence of F on the waiting time for values of the latter less than about 200 min. Furthermore, F varies quite substantially for a given waiting time depending on [ThC]/[ThB]. For values of the waiting time longer than 300 min, the variation of F with [ThC]/[ThB] is not very substantial (less than 10%). An almost linear relationship between F and the waiting time is observed for waiting times longer than about 400 min (see also Fig. 1 and 2).

Comparison of Fig. 5 and 6 show that for a given [ThC]/[ThB], F is lower for shorter sampling times than for longer sampling times. However, this only holds for waiting times of less than about 250 min and the trend reverses for waiting times longer than 250 min.

A rather interesting feature of Fig. 5 and 6 is that the F-factor becomes independent of the ratio [ThC]/[ThB] for a waiting time of about 215 min. This feature permits the determination of WL(Tn) by a single gross α-count without the need for assuming a particular thoron daughter disequilibrium ratio, as it is commonly done (25). The above procedure results in a more accurate determination of WL(Tn), and in a substantial reduction in the waiting time at which the α-count can be measured, i.e., from 300 min (25) to about 215 min.

From Equations 12 and 16 one can see that apart from a scaling factor, Fig. 5 and 6 represent the total α-activity collected on the filter versus

waiting time for several values of the ratio [ThC]/[ThB]. For values of [ThC]/[ThB]<1, the activity increases from time zero, it reaches a maximum value and then decreases with constant slope. The time at which the maximum activity is attained depends upon [ThC]/[ThB] and increases with increasing values of the latter ratio. For the extreme theoretical case of [ThC]/[ThB]=1, i.e., thoron daughter equilibrium, the maximum value of the activity takes place immediately after the end of sampling.

The behaviour of the α-activity on the filter indicated above can be understood as follows: For [ThC]=0, or [ThC]$<<$[ThB], the only α-activity on the filter is that originating from the decay of ThB. Thus, there will be an initial increase in the α-activity followed by a decay, after reaching a maximum, with an almost constant slope. The constant slope is due to the fact that an equilibrium is reached between [ThC] and [ThB] on the filter, after which the ThC decays with the half-life of ThB. In other words, the decay of ThC is solely determined by the presence and decay of its parent ThB. However, at equilibrium, i.e., [ThC]=[ThB], the α-activity on the filter is made up of two contributions: the decay of atmospheric ThC deposited on the filter and the decay of ThC originating from the decay of atmospheric ThB also deposited on the filter. The same applies for $0<$[ThC]/[ThB] $\bar{<} 1$.

The theoretical α-activity and experimental α-activity, determined from a thoron daughter calibration facility, versus waiting time is illustrated in Appendix A.

CONCLUSIONS

The F factor depends on both the waiting time after the end of the sampling period and the ratio [ThC]/[ThB], i.e., the thoron daughter disequilibrium ratio.

If α-activity measurements are conducted at least 300 min after the end of sampling, the F factor does not vary by more than 10% over the full range of theoretical values of the ratio [ThC]/[ThB], i.e., from 0 to 1.

The F factor is independent of the ratio [ThC]/[ThB] at approximately 215 min after the end of the sampling period. This feature can be used to determine WL(Tn) with more accuracy and at least 1.5 hr earlier than is commonly done using the one gross α-count method reported elsewhere.

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APPENDIX A

Figures A-1 and A-2 show, respectively, the theoretical α-activity in the filter (η=1, ε=1, Q=1 L/min, [ThB] = 1 pCi/L and [ThC]/[ThB] = 0.5) versus waiting time and the experimental α-activity versus waiting time. In the experimental case sampling was conducted in a thoron daughter calibration facility. Sampling and counting time were in both cases 5 min and 1 min, respectively.

It is not difficult to show that apart from a scaling factor to normalize experimental data, and notwithstanding some uncertainty in the experimental value of the ratio [ThC]/[ThB], Fig. A-1 and A-2 show good qualitative and quantitative agreement. These graphs have been reproduced here only for completeness.

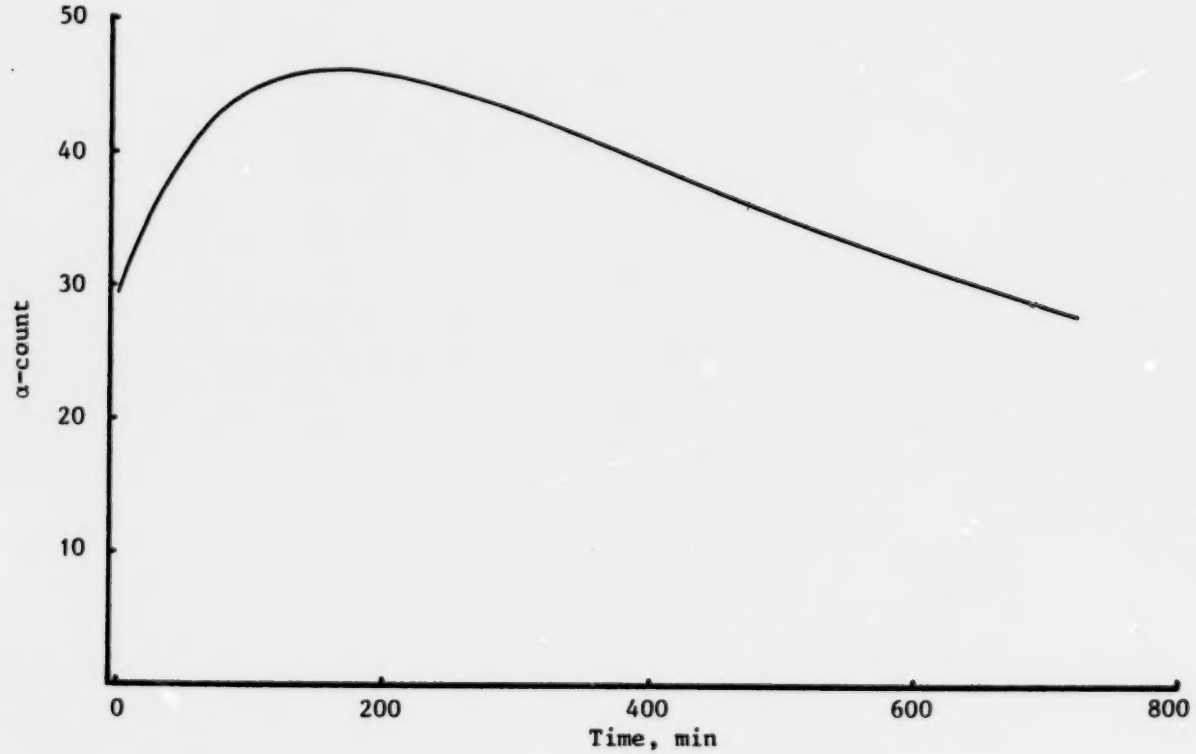

Fig. A-1 - Theoretical α-count versus waiting time for a [ThC]/[ThB] ratio of 0.5. Data normalized to Q = 1 L/min and ε = 1. Sampling time is 5 min, [ThB] = 1 pCi/L (37 mBq/L).

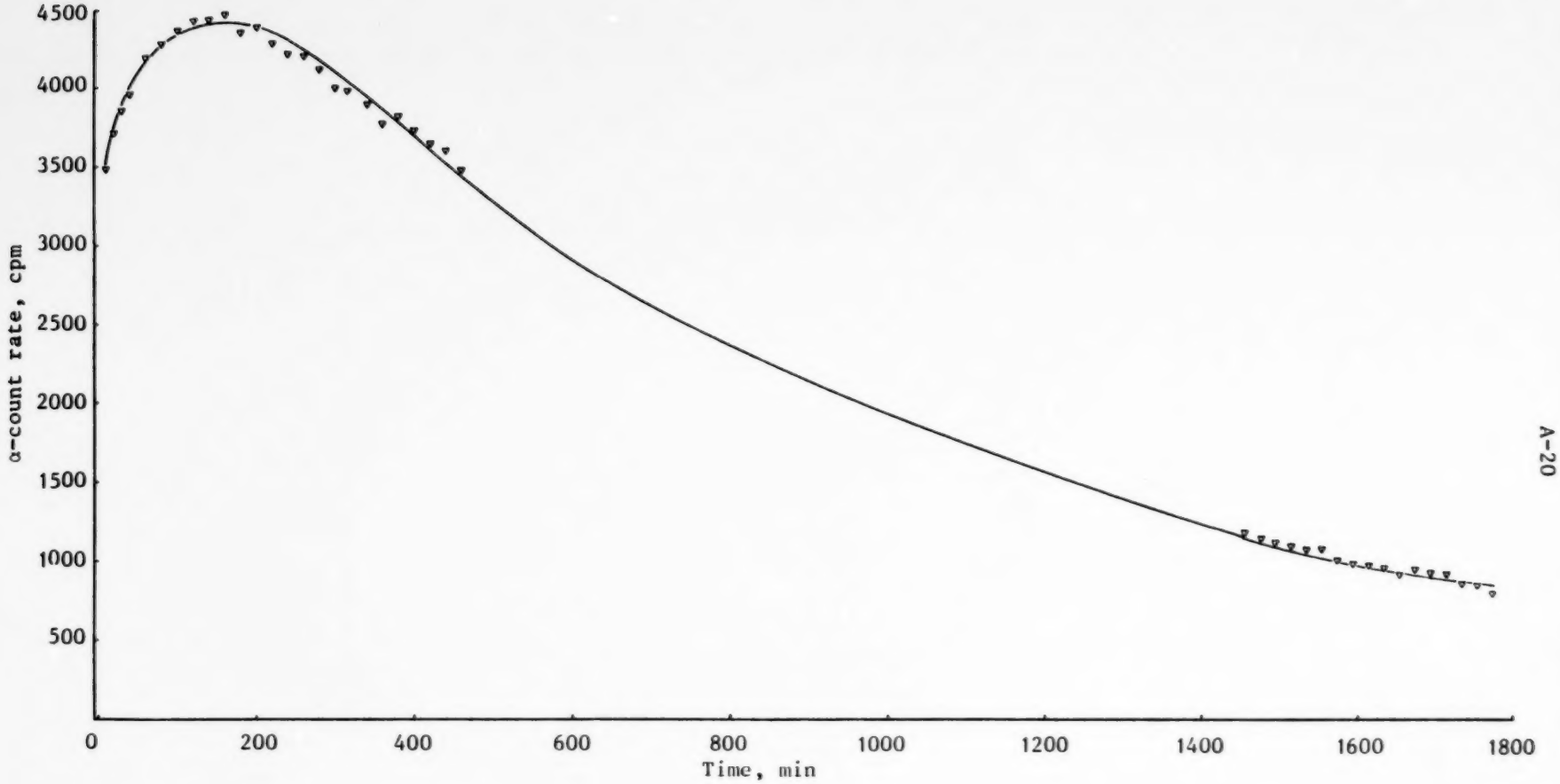

Fig. A-2 - Experimental α-count rate (cpm) versus waiting time (min). Sampling time is 5 min. [ThC]/[ThB] not known with certainty.